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This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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Additional inventors are being named on the <u>1</u> separately numbered sheets attached hereto					
TITLE OF THE INVENTION (500 characters max)					
Vesicles of Silicone Polyether Block Copolymers					
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[Page 1 of 2]

Respectfully submitted,

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Docket Number DC10103PSP1

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[Page 2 of 2]

Number 2 of 2

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Applicant(s): Shaow Burn Lin			DC10103 PSP 1	
Application No.	Filing Date	Examiner	Customer No.	Group Art Unit
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Invention: Vesicles of Silicone Polyether Block Copolymers

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**UNITED STATES PATENT APPLICATION
FOR**

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Vesicles
of
Silicone Polyether Block Copolymers

10

Having the inventor(s):

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Shaow Burn Lin
Kimmair Nguyen

20

25

FIELD OF THE INVENTION

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This application relates to vesicle compositions from $(AB)_n$ silicone polyether block copolymers, methods for preparing the vesicle compositions, and personal, household, and healthcare formulations containing the vesicle compositions.

BACKGROUND OF THE INVENTION

5 The aggregation behavior of surfactants in aqueous media is a robust area of investigation. Various vesicle compositions, such as liposomes and the like, have been developed for many applications with particular emphasis on the design of such systems for delivery of active materials via a pharmaceutical or personal care product formulation. Long-standing needs in this field are to identify vesicle compositions that form and entrap actives
10 easily, are stable under various chemical and mechanical stresses, and yet are able to deliver the actives in a controlled manner under desired conditions. To this end, many classes and types of surfactants have been investigated for their ability to form vesicles and thus entrap and deliver actives.

 Vesicles derived from silicone surfactants, and more particularly silicone polyether
15 surfactants, are of interest because of additional inherent benefits that this class of surfactants possesses vs other types. For example, silicone surfactants often have improved aesthetics in personal care formulations.

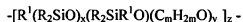
 The aggregation behavior of certain silicone polyethers has been discussed along with their ability to form vesicles. For example, US Patents 5,364,633 and 5,411,744 by Hill
20 teaches the self-assembly of certain silicone polyethers in aqueous media to form vesicles. PCT Application US2003/38455 by Lin teaches the entrapment of various oils in certain silicone polyether vesicles and their use in various personal care formulations.

 While these references represent advancements in the art, a need still exists to create silicone vesicles having improved stability. For example, the silicone vesicles formed from
25 the self-assembly procedures can lack durability. This lack of durability can limit the processing conditions they can be subject to in the formation of various finished products. Furthermore, such self-assembled silicone vesicles can lack the ability to provide sustained release of loaded actives over an extended period of time. Thus, a need exists for a process to prepare silicone vesicles having improved durability and the ability to release loaded actives
30 over an extended period of time.

The present inventors have discovered that a particular-class of silicone polyethers, namely $(AB)_n$ block copolymer silicone polyethers, form vesicle compositions in aqueous media. The vesicle compositions possess enhanced stability and are useful to prepare formulations for the delivery of personal, household, and healthcare active materials.

SUMMARY OF THE INVENTION

- 5 The present invention relates to vesicle compositions comprising a $(AB)_n$ block silicone polyether copolymer. Preferably the $(AB)_n$ block silicone polyether copolymer is a polyorganosiloxane-polyoxyalkylene block copolymer having the average formula;



- 10 where

x and y are greater than 4, m is from 2 to 4 inclusive, z is greater than 6,

R is independently a monovalent organic group,

R^1 is a divalent organic group containing 2 to 30 carbons.

- 15 The present invention further relates to a process for making a vesicle composition comprising;

I) mixing,

A) a $(AB)_n$ silicone polyether copolymer,

B) an optional water miscible volatile solvent,

- 20 C) water,

to form an aqueous dispersion of the $(AB)_n$ silicone polyether copolymer,

II) optionally, removing the water miscible volatile solvent from the aqueous dispersion to form the vesicle composition.

- 25 Furthermore, the present invention relates to personal, household, and healthcare formulations containing the vesicle compositions.

DETAILED DESCRIPTION OF THE INVENTION

The vesicle compositions of the present invention comprise a (AB)_n block silicone polyether copolymer. As used herein, "vesicle" is the common art term referring to entities formed from the aggregation of surfactant and/or lipid molecules in aqueous medium, characterized by the presence of one or more walls or membranes formed from a surfactant and/or lipid compound creating one or more internal voids. The walls or membranes formed by the surfactant and/or lipid in vesicles can be unilamellar, bilamellar, oligolamellar, or multilamellar structures. The formation of vesicles can be confirmed by techniques common in the state of the art. Typically, the lamellar phase structure in vesicles exhibit birefringence when examined with a cross polarizing microscope. Alternatively, the formation of vesicles can be demonstrated by Cyro-Transmission Electron Microscopy (Cryo-TEM) techniques.

The vesicle compositions of the present invention can comprise any (AB)_n block silicone polyether copolymer having sufficient amphiphilic or surfactant character to allow the structuring of the silicone polyether copolymer in aqueous media for vesicle formation.

Preferably the (AB)_n block silicone polyether copolymer of the vesicle composition is a polyorganosiloxane-polyoxyalkylene block copolymer having the average formula;



where x and y are greater than 4, m is from 2 to 4 inclusive, z is greater than 2, R is independently a monovalent organic group containing 1 to 20 carbons, R¹ is a divalent hydrocarbon containing 2 to 30 carbons.

The siloxane block in Formula I is a predominately linear siloxane polymer having the formula (R₂SiO)_x, wherein R is independently selected from a monovalent organic group, x is a integer greater than 4, alternatively x ranges from 20 to 100, or from 30 to 75.

The organic groups represented by R in the siloxane polymer are free of aliphatic unsaturation. These organic groups may be independently selected from monovalent hydrocarbon and monovalent halogenated hydrocarbon groups free of aliphatic unsaturation.

- These monovalent groups may have from 1 to 20 carbon atoms, alternatively 1 to 10 carbon atoms, and are exemplified by, but not limited to alkyl groups such as methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl; and halogenated hydrocarbon groups such as 3,3,3-trifluoropropyl, 3-chloropropyl, and dichlorophenyl. At least 50 percent, alternatively at least 80%, of the organic groups in the organopolysiloxane may be methyl (denoted as Me). Typically, the siloxane block is a predominately linear polydimethylsiloxane having the formula $(\text{Me}_2\text{SiO})_x$, where x is as defined above.

- The polyoxyalkylene block of the silicone polyether is represented by the formula $(\text{C}_m\text{H}_{2m}\text{O})_y$, wherein m is from 2 to 4 inclusive, and y is greater than 4, alternatively y can range from 5 to 30, or alternatively from 5 to 22. The polyoxyalkylene block typically can comprise oxyethylene units $(\text{C}_2\text{H}_4\text{O})_y$, oxypropylene units $(\text{C}_3\text{H}_6\text{O})_y$, oxybutylene units $(\text{C}_4\text{H}_8\text{O})_y$, or mixtures thereof. Typically, the polyoxyalkylene block comprises oxyethylene units $(\text{C}_2\text{H}_4\text{O})_y$.

- At least one end of each polyoxyalkylene block in Formula I is linked to a siloxane block by a divalent organic group, designated R^1 . This linkage is determined by the reaction employed to prepare the $(\text{AB})_n$ block silicone polyether copolymer. The divalent organic groups of R^1 may be independently selected from divalent hydrocarbons containing 2 to 30 carbons and divalent organofunctional hydrocarbons containing 2 to 30 carbons.
- Representative, non-limiting examples of such divalent hydrocarbon groups include; ethylene, propylene, butylene, pentylene, hexylene, heptylene, octylene, and the like. Representative, non-limiting examples of such divalent organofunctional hydrocarbons groups include acrylate and methacrylate. Typically, R^1 is propylene, $(-\text{CH}_2\text{CH}_2\text{CH}_2-)$.

- The $(\text{AB})_n$ block silicone polyethers are endblocked. The endblocking unit is also determined by the reaction employed to prepare the $(\text{AB})_n$ block silicone polyether copolymer, which is generally the residual reactive groups of the reactants used. For example, the $(\text{AB})_n$ block silicone polyether copolymers can be prepared by the metal catalyzed hydrosilylation reaction of a diallyl polyether (i.e. an allyl group is present on each molecular terminal end) with a SiH terminated polyorganosiloxane. The resulting $(\text{AB})_n$ block silicone polyether copolymer would have polyoxyalkylene blocks linked to the silicone blocks via a propyleneoxy group $(-\text{CH}_2\text{CH}_2\text{CH}_2\text{O}-)$, and using a slight molar excess of the

allyl polyether would result in an allyl endblock unit ($-\text{CH}_2\text{CHCH}_2$). Alternative endblock units can result from the addition of other molecules in the reaction employed to prepare the $(\text{AB})_n$ block silicone polyether copolymer that are capable of reacting with the siloxane or polyether block intermediates. For example, the addition of organic compounds having

5 mono-terminated aliphatic unsaturation (such as a mono allyl terminated polyether) will result in the endcapping of the $(\text{AB})_n$ block silicone polyether copolymer with that organic compound. Preferably, the endblocking units of the $(\text{AB})_n$ block silicone polyether is an allyl ether ($\text{CH}_2=\text{CHCH}_2\text{O}-$) or allyl polyether.

The molecular weights of the $(\text{AB})_n$ block silicone polyether copolymers will be

10 determined by the number of repeating siloxane and polyoxyethylene blocks, as indicated by the subscript z in Formula I. Typically, the value of z is such to provide weight average molecular weights (M_w) to range from 1,500 to 150,000, alternatively, from 10,000 to 100,000.

The $(\text{AB})_n$ SPEs of the present vesicle compositions have a molar ratio of the total

15 siloxane units to the polyoxyethylene units in the $(\text{AB})_n$ block silicone polyether. This molecular parameter is expressed by the value of $x/(x+y)$ in Formula I. The value of $x/(x+y)$ can vary from 0.4 to 0.9, or alternatively from 0.55 to 0.9.

The $(\text{AB})_n$ SPEs useful to prepare the vesicle compositions of the present invention can be prepared by any method known in the art for preparing such block copolymers.

20 Typically however, the $(\text{AB})_n$ SPEs useful in the preparation of the vesicle compositions of the present invention are obtained from a method comprising reacting an SiH terminated organopolysiloxane with a polyoxyethylene having an unsaturated hydrocarbon group at each molecular terminal, in a hydrosilylation reaction, wherein the mole ratio of the unsaturated hydrocarbon groups to SiH in the reaction is at least 1:1.

25 The vesicle compositions of the present invention can be prepared by mixing the $(\text{AB})_n$ SPEs with water using any technique known in the state of the art for creating vesicle compositions. The type and extent of the mixing technique will depend on the specific structure of the $(\text{AB})_n$ SPE chosen. For example, some $(\text{AB})_n$ SPEs will form vesicle compositions spontaneously when mixed with water, while others $(\text{AB})_n$ SPEs will require

30 the presence of a water soluble solvent to facilitate the formation of vesicles. In a preferred

embodiment, the vesicle compositions of the present invention are prepared using a process comprising;

I) mixing,

A) a $(AB)_n$ silicone polyether copolymer,

5 B) a water miscible volatile solvent,

C) water,

to form an aqueous dispersion of the $(AB)_n$ silicone polyether copolymer,

II) optionally, removing the water miscible volatile solvent from the aqueous dispersion to form the vesicle composition.

- 10 The $(AB)_n$ SPEs, component A), is the same as described above. Component B) is a water-miscible volatile solvent. As used herein "water-miscible" means the solvent forms a dispersion with water at room temperature for at least several hours. "Volatile" means the solvent has a higher vapor pressure than water at various temperatures. As such, when the aqueous dispersion of the organopolysiloxane and solvent are subjected to conditions to
- 15 remove the solvent, such as heating the dispersion under reduced pressures, the solvent is primarily removed first, allowing all or most of the water to remain in the composition.

- Suitable water-miscible volatile solvents for vesicle dispersion preparation include organic solvents such as alcohols, ethers, glycols, esters, acids, halogenated hydrocarbons, diols. The organic solvents should be miscible with water at the proportion and lower in order
- 20 to effectively disperse silicones and maintain stable and uniform dispersion overtime. For the purpose of illustration, water-miscible alcohols include methanol, ethanol, propanol, isopropanol, butanol, and higher hydrocarbon alcohols; ethers include glycol ethers, methyl-ethyl ether, methyl isobutyl ether (MIBK), etc; glycols include propylene glycols, esters include esters of triglycerol, the esterification products of acid and alcohol; halogenated
- 25 hydrocarbons include chloroform. Typically water-miscible organic solvents are solvents with relatively low boiling points ($<100^\circ\text{C}$) or high evaporation rate, so they may be removed under vacuum with ease. The most preferred water-miscible organic solvents for this invention are volatile alcohols including methanol, ethanol, isopropanol, and propanol. These alcohols can be removed from aqueous mixtures containing silicone vesicle dispersions via
- 30 vacuum stripping at ambient temperature.

The order of mixing components A), B), and C) is not critical, but typically A) and B) are first mixed and then water added to the mixture. There are no special requirements or conditions needed for effecting the mixing of components A), B), and C). The mixing can be conducted in a batch, semi-continuous, or continuous process.

- 5 The amount of components A), B), and C) can vary in the process, but typically range as follows;

A) 2 to 50 wt%, alternatively 2 to 25 wt %, or alternatively 2 to 15 wt%,

B) 0 to 50 wt%, alternatively 2 to 30 wt %, or alternatively 2 to 20 wt%,

C) sufficient amount to provide the sum of the wt% of A), B), and C) to equal 100%

- 10 The amount of B) water-miscible volatile solvent used to disperse the $(AB)_n$ SPEs depends on the type of organopolysiloxane and how much hydrophilic groups are present. Typically, the aqueous mixture to effectively disperse silicones comprises of 5 to 80 parts of solvent and 20 to 95 parts of water; alternatively 5 to 50 parts of water, or alternatively 10 to 40 parts water.

- The formation of vesicles in the compositions of the present invention can be
15 confirmed by techniques common in the state of the art. Typically, vesicles having a lamellar phase structure which exhibit birefringence when examined with a cross polarizing microscope. Alternatively, the formation of vesicles can be demonstrated by Cryo-Transmission Electron Microscopy (Cryo-TEM) techniques. Particle size measurements can also be used to indicate that the organopolysiloxanes are sufficiently dispersed in aqueous
20 medium typical of vesicle sizes. For example, average particle sizes of less than 0.500 μm (micrometers), are typical for dispersed vesicles. Vesicles having a average particle size of less than 0.200 μm , or 0.100 μm are possible with the teachings of the present invention.

- The present invention also relates to vesicle compositions further comprising a personal, household, or health care ingredient. Thus, the vesicle compositions can be used to
25 entrap, and subsequently deliver after application, a personal, household care, or health care ingredient. A listing of possible personal, household, or health care ingredients is taught in WO 03/101412, which is incorporated herein by reference. The personal or health care ingredient can also be selected from a personal or health care "active", that is, any compound known to have either cosmetic and/or pharmaceutical activity. A representative listing of
30 such personal or health care actives are disclosed in US Patent 6,168,782, which is hereby incorporated by reference.

Compositions prepared according to the invention can be used in various over-the-counter (OTC) personal care compositions, health care compositions, and household care compositions, but especially in the personal care arena. Thus, they can be used in antiperspirants, deodorants, skin creams, skin care lotions, moisturizers, facial treatments
5 such as acne or wrinkle removers, personal and facial cleansers, bath oils, perfumes, colognes, sachets, sunscreens, pre-shave and after-shave lotions, liquid soaps, shaving soaps, shaving lathers, hair shampoos, hair conditioners, hair sprays, mousses, permanents, depilatories, hair cuticle coats, make-ups, color cosmetics, foundations, blushes, lipsticks, lip balms, eyeliners, mascaras, oil removers, color cosmetic removers, nail polishes, and
10 powders.

EXAMPLES

The following examples are presented to further illustrate the compositions and methods of this invention, but are not to be construed as limiting the invention. All parts and percentages in the examples are on a weight basis and all measurements were obtained at 23°C, unless indicated to the contrary.

Materials

The representative (AB)_n silicone polyethers, herein designated as (AB)_n SPE, used in the vesicle compositions of the present invention were prepared using the following starting materials.

MD_xM' siloxanes – a series of dimethyl-hydrogen terminated (Me₂HSiO) linear polydimethylsiloxanes of varying degree of polymerizations (as designated by x) were prepared using well known siloxane polymerization techniques. The average formulas for this series of siloxanes are summarized in Table 1.

Allyl terminated polyethers – a series of alpha, omega-diallyloxy polyethers having the average formula (CH₂=CHCH₂O(CH₂CH₂O)_mCH₂CH=CH₂) were used to prepare the (AB)_n silicone polyether block copolymers.

Polyglycol AA600, AA1200, and AA2000, used as obtained from Clariant (Mt. Holly, NC), contained on average 6, 12, and 22 ethylene oxide units (designated as EO) (ie. m = 6, 12, and 22)

Testing Procedures

Particle Size

5 *Cryo-Transmission Electron Microscopy (TEM)*

The vesicle compositions were analyzed via Cryo-TEM techniques according to the following procedure. Around 2.3 μl of aqueous sample solution was loaded using a micropipette on a lacey carbon film coated Cu TEM grid that was cleaned and rinsed with acetone and chloroform. The samples were diluted to 5% solution with de-ionized water. The excess fluid on the grid surface was removed by blotting the surface with a filter paper for 1.5 second to make an aqueous thin film for TEM. The grid was then plunged into a liquid ethane contained in a small vessel located in a larger liquid nitrogen vessel under $-175\text{ }^{\circ}\text{C}$ atmosphere in the cryo-plunge system to vitrify the water film on the grid and to avoid water crystallization. The quenched sample grid was transferred in to the cryo-grid box in the cryo-plunge system. The grid box containing the sample was transferred into a Gatan cryo-transfer system filled with liquid nitrogen and loaded in a cryo-TEM stage, which has been positioned in the cryo-transfer system and cooled down to below $-160\text{ }^{\circ}\text{C}$. The sample was loaded in TEM (JEOL 2000FX) and the images were observed at below $-160\text{ }^{\circ}\text{C}$. A much colder finger, cooled to $-180\text{ }^{\circ}\text{C}$ in TEM using liquid nitrogen, was present to reduce any possible contamination on the cold specimen surface under high vacuum during TEM analysis. The digital images, as shown herein, were taken using a Gatan CCD camera attached at the bottom of the TEM column and Digital Micrograph software.

Examples 1 – 6 (Reference)

Various (AB)_n SPEs, as summarized in Tables 1 were prepared via the platinum catalyzed hydrosilylation of the SiH siloxanes with the allyl polyethers utilizing the following
5 general procedure.

Procedure to Prepare SPEs

A 1000 ml three neck round bottom flask equipped with temperature probe, electrical stirrer, and condenser was charged with an amount (as indicated in Table 1) of a polyethylene
10 glycol diallyl ether (Clariant Corp., Mt. Holly, NC), 61 gram of xylene and 0.28 gram of sodium acetate. The contents of the flask were then heated to 100 °C. A dimethylhydrogen endblocked polydimethyl siloxane was added dropwise via an addition funnel (amount and structures shown in Table 2). After adding 5 gram of the siloxane, 0.60 gram of platinum catalyst (1,3-diethenyl-1,1,3,3-tetramethyldisiloxane platinum complex in dimethyl siloxane)
15 was added to the mixture. When half of the siloxane was added, an additional 0.69 gram of Pt catalyst was added, followed by 0.71 gram of Pt when all the siloxane addition was complete. The reaction mixture was allowed to mix for 1 hour for the polymer to grow. The xylene solvent was then removed via vacuum stripping at 150 °C.

Multiple batches of (AB)_n SPEs block copolymers were made, in some cases, to
20 create molecular weight variations of the same (AB)_n SPEs block copolymers from a given siloxane and polyether combinations. These also demonstrate the suitability of (AB)_n copolymers having different chain lengths (i.e. n values) to prepare the vesicle compositions.

Table 1

SPE Example Reference #	Siloxane block	Siloxane used (g)	Polyether used	Polyether used (g)	M _w
(AB) _n SPE 1	M'D ₃₀ M'	180.7	AA1200	119.33	19,486
(AB) _n SPE 2	M'D ₅₀ M'	394.3	AA1200	105.78	50,108
(AB) _n SPE 3A	M'D ₇₅ M'	850.1	AA1200	149.26	40,158
(AB) _n SPE 3B	M'D ₇₅ M'	850.1	AA1200	180.0	44,885
(AB) _n SPE 4	M'D ₁₀₀ M'	223.1	AA1200	25.0	88,800
(AB) _n SPE 5	M'D ₅₀ M'	161.4	AA2000	88.7	22,306

Examples 12 – 16***Vesicle Compositions from (AB)_n SPE 2***

5 The following procedure was used to prepare the vesicle compositions summarized in Table 3 as Examples 12 and 13.

Isopropanol (IPA) was added to (AB)_n SPE 2, (a (AB)_n block copolymer of M'D₅₀M' siloxane and polyglycol AA1200 polyether, having a weight-average molecular weight Mw of 50,108 g/mole), to provide a homogeneous mixture. With continuous mixing, water was
10 added slowly to form a homogeneous dispersion having an average particle size of 0.208 μm. The IPA in the dispersion was then removed using a Rotovapor under vacuum at ambient temperature, to yield an alcohol-free, homogeneous dispersion having an average particle size of 0.223 μm, designated as Example 13.

Three additional vesicle compositions using (AB)_n SPE 2 block copolymer were
15 made, following the procedure of Example 12, except an optional homogenization step was introduced after the mixture was made and before the vacuum strip. Ethanol (EtOH) was used in place of IPA as the alcohol. The compositions are summarized in Table 3. As the data indicates, the homogenization step reduced the average particle size and maintained the homogeneity of the dispersion. Removal of the volatile alcohol (EtOH) did not compromise
20 the quality of the dispersion.

The particle size distributions of the compositions for Example 14 – 16 are shown in figure 1. The cyro TEM images of the compositions of Examples 13 – 16, as shown in Figures 2 – 5, confirm the presence of the vesicle structures.

Table 3

Example # Experiment #	12	13	14	15	16
Process History	Mixed	Mixed, then stripped	Mixed	Mixed, homogenized	Mixed, homogenized, then stripped
Alcohol type	IPA	IPA	EtOH	EtOH	EtOH
(AB)n SPE 2, g	30.27	22.35	30.25	30.25	16.30
Alcohol, g	90.41	66.73	90.98	90.98	49.01
Water	180.78	133.44	180.06	180.06	97.00
Batch size, as mixed	301.46	222.52	301.29	301.29	162.31
Alcohol Removed		-62.40			-46.90
Batch size, after strip		160.12			115.41
Final composition					
Wt. % Polymer	10.0	14.0	10.04	10.04	14.12
Wt. % alcohol	30.0	2.7	30.20	30.20	1.83
Wt. % Water	60.0	83.3	59.76	59.76	84.05
Avg. particle size, μm	0.2081	0.2235	0.387	0.1388	0.175
D(v, 0.5), μm	0.1003	0.085	0.2556	0.1023	0.1474
D(v, 0.9), μm	0.552	0.828	0.912	0.2872	0.321

Examples 17–19***Vesicle Compositions from (AB)_n SPE 1***

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Vesicle compositions of (AB)_n SPE 1 an (AB)_n block copolymer of M'D₃₀M' siloxane and polyglycol AA1200 polyether, having a weight-average molecular weight Mw of 19486 g/mole were prepared following the procedure of Examples 12-13. These vesicle compositions are summarized in Table 4 as Examples 17 – 19. All three compositions had an average particle size distribution of less than 40 nm (0.040 μm). These examples demonstrate that the removal of alcohol did not affect the quality of dispersion and the homogenization step is optional.

15

Table 4

Example #	17	18	19
Process History	Mixed	Mixed, homogenized	Mixed, homogenized, stripped
19162-20 (AB) _n SPE 1, g	30.04	30.040	18.13
EtOH, g	90.41	90.410	54.40
Water, g	181.02	181.020	108.80
Batch size, as mixed, g	301.47	301.47	181.34
EtOH Removed			-66.6
Batch size, after strip, g			114.7
Final dispersion composition			
Wt. % SPE	10.0	10.0	15.8
Wt. % EtOH	30.0	30.0	0.0
Wt. % Water	60.0	60.0	84.2
Appearance	Almost clear dispersion	Almost clear dispersion	Hazy uniform dispersion
Avg. particle size, μm	0.023	0.039	0.030
D(V, 0.5), μm	0.022	0.027	0.023
D(v, 0.9), μm	0.034	0.080	0.057

Examples 20 – 23*Vesicle Compositions from (AB)_n SPE 3 A & B*

- 5 Vesicle compositions were prepared from (AB)_n SPE 3 A and B, (AB)_n silicone polyether block copolymers of M'D₇₅M' siloxane and polyglycol AA1200 polyether having a weight-average molecular weight Mw of 40, 158 and 44,885 g/mole, respectively. These dispersions were made following the procedure described in Examples 12 – 13 and are summarized in Table 5.

10

Table 5

ID example	20	21	22	23
Process History	Mixed	Mixed	Mixed	Mixed
19162-56 (AB) _n SPE, g	2.022	2.038		
19162-57 (AB) _n SPE, g			2.092	2.097
IPA, g	9.082		9.011	
EtOH, g		9.079		9.065
Water, g	9.016	9.123	9.096	9.106
Dispersion Composition				
Wt. % SPE	10.0	10.1	10.4	10.3
Wt. % Alcohol	45.1	44.9	44.6	44.7
Wt. % Water	44.8	45.1	45.0	44.9
Appearance	Cloudy, homogeneous dispersion	Cloudy, homogeneous dispersion	Cloudy, homogeneous dispersion	Cloudy, homogeneous dispersion
Avg. particle size, um	0.740	0.167	1.715	0.189
D(v, 0.5), um	0.660	0.138	1.637	0.142
D(v, 0.9), um	1.642	0.311	2.655	0.396

Examples 24 – 25*Alcohol free vesicle compositions from (AB)_n SPE 3 B*

- Alcohol-free vesicle compositions were prepared from (AB)_n SPE 3 B an (AB)_n silicone polyether block copolymer of M'D₇₅M' siloxane and polyglycol AA1200 polyether, having a weight-average molecular weight Mw of 44,885 g/mole following the procedure of Examples 12 –13, and removing the alcohol under reduced pressure. The compositions are summarized in Table 6.

Table 6

ID example	24	25
Process History	Mixed	Mixed, stripped
(AB) _n SPE 3B, g	30.820	27.783
IPA, g	120.470	108.597
Water, g	150.980	136.100
Batch size, as mixed	302.27	272.48
IPA Removed		-100.5
Batch size, stripped		172.0
Dispersion composition		
Wt. % SPE	10.20	16.15
Wt. % IPA	39.86	4.71
Wt. % Water	49.95	79.14
Appearance	Hazy to slight cloudy; homogeneous	Milky opaque, uniform dispersion
Avg. particle size, um	0.604	0.784
D(v, 0.5), um	0.523	0.397
D(v, 0.9), um	1.064	0.779

Example 26 - 27*Vitamin A palmitate loaded in vesicles from (AB)_n SPE 1*

- 5 Vitamin A palmitate was first mixed with isopropanol at 50/50 ratio. The vitamin / IPA mixture was then mixed with (AB)_n SPE 1, a (AB)_n block copolymer of M'D₃₀M' siloxane and polyglycol AA1200 polyether, having a weight-average molecular weight Mw of 19486 g/mole to homogenous. Ethanol was then admixed to form a homogeneous mixture. While under continuous mixing, de-ionized water was slowly and gradually incorporated into
- 10 the SPE / vitamin / alcohol mixture, till homogenous. The mixture was homogenized, using an APV-2000 Gaulin homogenizer, producing a homogeneous dispersion with sub-micron particle size, identified as Example 26 in Table 8. The Example 26 composition was then further homogenized. The alcohol was removed under reduced pressure at ambient temperature to produce a composition having an average particle size of 0.54 um, listed as
- 15 Example 27 in Table 8. The removal of alcohol processing aid did not affect the dispersion quality and the particle size.

Table 8

Example #	26	27
Process History	Mixed, homogenized	Mixed, homogenized, and stripped
Vitamin A palmitate, g	6.65	4.69
IPA, g	6.65	4.58
EtOH, g	90.86	62.88
(AB)n SPE 1, g	30.17	20.88
D.I. Water, g	180.15	124.65
Batch size, as mixed	314.48	217.62
Alcohol removed, g		-60.1
Batch size, after strip		157.52
Final dispersion composition		
Wt. % VAP	2.11	2.91
Wt. % Polymer	9.59	13.25
Wt. % Alcohol	31.05	4.73
Wt. % Water	57.28	79.11
Appearance	Milky yellow liquid	Milky yellow liquid
Avg. particle size, μm	0.540	0.574
D(v, 0.5), μm	0.358	0.353
D(v, 0.9), μm	1.231	1.266

Example 28

- The following vitamin A palmitate loaded (AB)_n SPE vesicles in water dispersion was prepared according to the method shown in the previous examples of this invention. The (AB)_n SPE is a copolymer of 50 dp siloxane and Polyglycol AA1200 polyether. The final composition of the vesicle dispersion is shown in Table 9.

Table 9

Example # 28	
(AB) _n SPE, g	118.00
Vitamin Premix	35.38
Vit A Palmitate in the premix, g	25.16
D.I. Water, g	346.62
Wt. % SPE Polymer	23.60
Wt. % VAP	5.03
Wt. % Tocopherol	0.51
Wt. % Silicone fluid	1.28
Wt. % Water	69.32
Appearance	Beige, milky dispersion, smooth
pH of the vesicle dispersion	5.39
Avg. vesicle size, μm	0.2583

10

The vitamin-loaded SPE vesicles are easy to formulate into skin care formulations. Oil-based vitamins can be easily incorporated into water-based formulations. The following examples provide such illustrations.

15

Example 29*Oil-in-water body lotion*

Ingredients	Parts
<u>Part A</u>	
Cetearyl Alcohol	3
Diisopropyl Adipate (Crodamol DA)	5
Dimethicone (Dow Corning Silicone 200 / 100 cstks)	0.5
Potassium cetyl phosphate	1.5
Buthylated hydroxytoluene	0.05
Cheating agent (EDTA)	0.1
Phenoxyethanol	0.6
<u>Part B</u>	
Water	up to 100
Carbomer 980 thickener	30
Potassium hydroxyde	1.5
<u>Part C</u>	
Vitamin A palmitate loaded SPE vesicles	19.88

5

To prepare the body lotion, the following procedure was followed: The ingredients in Part A were mixed and heated to 85 °C to homogeneous. Cool the part A mixture to 40 °C, then incorporate the part B ingredients. Cooled the mixture to ambient temperature. Incorporate vitamin A palmitate-loaded SPE vesicles into the mixture and mix to homogeneous. The final mixture is a smooth, slightly yellowish creamy lotion.

10

Example 30*Simple Moisturing Gel for Skin*

- 5 The (AB)_n type SPE vesicles can be easily formulated into aqueous based gel formulations. SPE vesicles provide a convenient means to incorporate oil-soluble vitamins into water-rich gel formulations.

Ingredients	Parts
<u>Part A</u>	
Water	to 100%
Preservative	0.30%
Polyacrylamide, C13-14 Isoparaffin, laureth-7 (Sepigel 305)	1%
<u>Part B</u>	
Vitamin A palmitate-loaded SPE vesicles	19.88

- 10 To prepare the gel, the following procedure was followed: The ingredients in Part A were mixed to homogeneous. Vitamin A palmitate-loaded SPE vesicles dispersion was then incorporated and mixed to homogeneous. The final product is a beige, smooth gel.

- 15 To further demonstrate the integrity of vesicles in formulations, cryo-TEM images of the “as formulated” products were taken. An image of the gel from the above example is shown figure 6. As illustrated, the vesicles and aggregates of vesicles were well preserved.

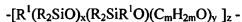
- 20 A cryo-TEM image of the “as prepared” body lotion illustrated in Example A, prepared from (AB)_n SPE vesicle is shown in figure 7. The characteristic vesicles and aggregate structures uniquely associated with the (AB)_n SPE vesicles shown in figure 7.

CLAIMS

1. A vesicle composition comprising a (AB)_n block silicone polyether copolymer.

5

2. The vesicle composition of claim 1 wherein the (AB)_n block silicone polyether copolymer is a polyorganosiloxane-polyoxyalkylene block copolymer having the average formula;



10 where

x and y are greater than 4, m is from 2 to 4 inclusive, z is greater than 2,

R is independently a monovalent organic group,

R¹ is a divalent hydrocarbon containing 2 to 30 carbons.

15 3. The vesicle composition of claim 2 wherein m is 2.

4. The vesicle composition of claim 2 wherein x ranges from 20 to 100.

5. The vesicle composition of claim 2 wherein R is methyl and R¹ is propylene.

20

6. The vesicle composition of claim 2 wherein the polyorganosiloxane-polyoxyalkylene block copolymer has a weight average molecular weight ranging from 1,500 to 150,000.

7. The vesicle composition of claim 2 wherein the value of x/(x+y) ranges from 0.4 to 0.9.

25

8. A process for making a vesicle composition comprising;

I) mixing,

A) a $(AB)_n$ silicone polyether copolymer,

B) an optional water miscible volatile solvent,

C) water,

to form an aqueous dispersion of the $(AB)_n$ silicone polyether copolymer,

II) optionally, removing the water miscible volatile solvent from the aqueous dispersion to form the vesicle composition.

9. The process of claim 8 wherein the $(AB)_n$ silicone polyether copolymer is a polyorganosiloxane-polyoxyalkylene block copolymer having the average formula;



where

x and y are greater than 4, m is from 2 to 4 inclusive, z is greater than 2,

R is independently a monovalent organic group,

R^1 is a divalent hydrocarbon containing 2 to 30 carbons.

10. The vesicle composition produced by the process of claim 9.

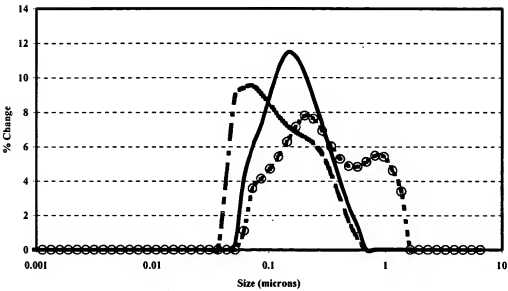
11. The vesicle composition of claim 1 or 9 further comprising a personal, household, or healthcare active ingredient.

12. A personal, household, and healthcare composition comprising the vesicle composition of claims 1, 10 or 11.

ABSTRACT

- 5 Vesicle compositions from $(AB)_n$ silicone polyether block copolymers, methods for preparing the vesicle compositions, and personal, household, and healthcare formulations containing the vesicle compositions are disclosed.

Figure 1

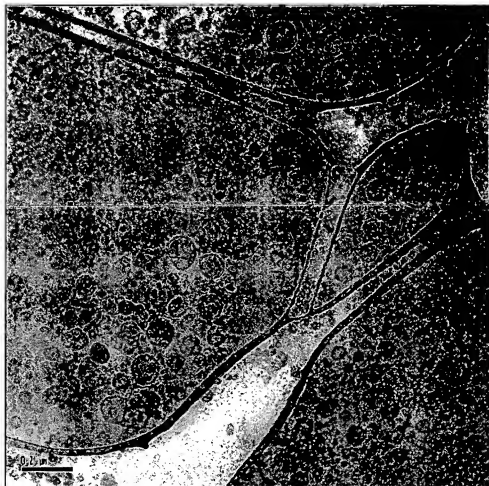


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Particle size distributions for vesicle compositions, Examples 14 - 16

Figure 2
(AB)_n SPE 2 in water dispersion

5

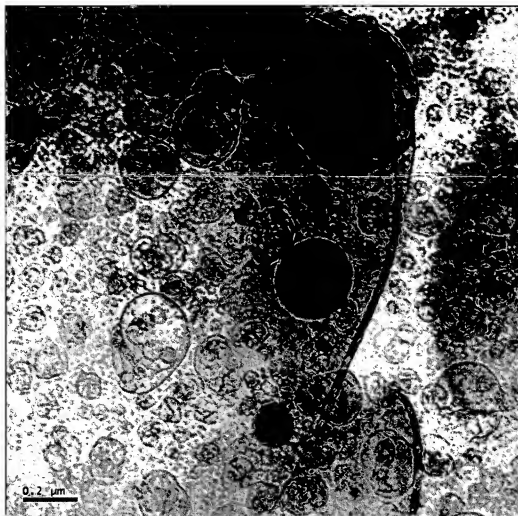


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Figure 3

(AB)_n SPE Block Copolymer Dispersion:
As Dispersed in 10 SPE /30EtOH / 60 Water

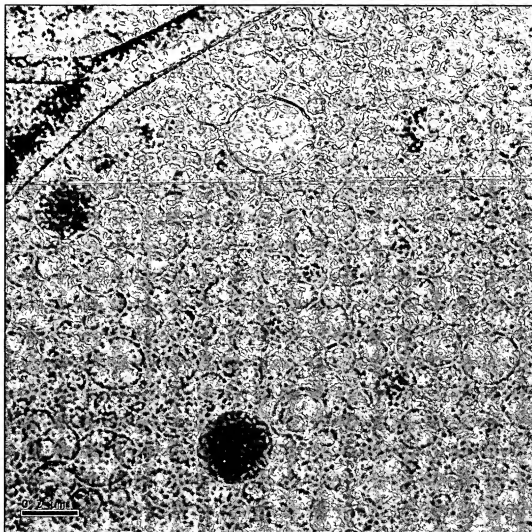
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Figure 4
(AB)_n SPE Block Copolymer Dispersion:
Mixed, Homogenized in EtOH / Water

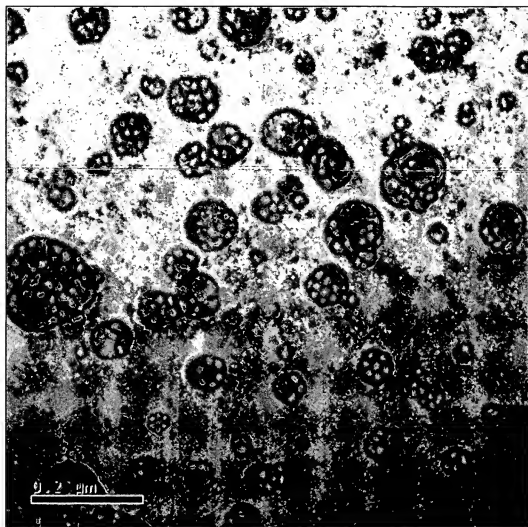
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Figure 5
(AB)n SPE 1 Dispersion:
Stripped to Remove EtOH

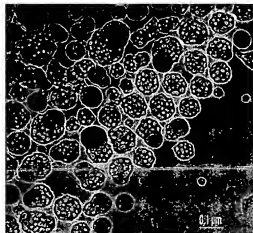
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Figure 6

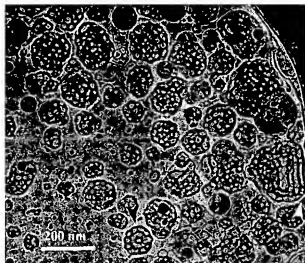
Cryo-TEM image of body lotion of Example 29
formulated from vitamin-loaded SPE vesicles



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Figure 7

Cryo-TEM image of gel from Example 30
formulated from vitamin-loaded SPE vesicles



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